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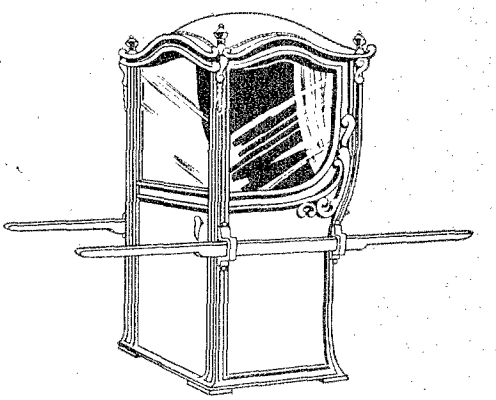

Release and Movement of Radionuclides in Soils Contaminated with Fallout Materials from an Underground Thermonuclear Detonation

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UCLA SCHOOL OF MEDICINE

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RELEASE AND MOVEMENT OF RADIONUCLIDES IN SOILS CONTAMINATED WITH FALLOUT MATERIAL FROM AN UNDERGROUND THERMONUCLEAR DETONATION

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ABSTRACT

Fallout material from an underground thermonuclear detonation was analyzed to determine the presence of several longer lived radionuclides. The following radionuclides were identified: Zr^{95}-Nb^{95}, Ru^{103}, Ru^{106}-Rh^{106}, I^{131}, Cs^{137}-Ba^{137}, Ba^{140}-La^{140}, Ce^{141}, Ce^{144}-Pr^{144}, Sc^{46}, Mn^{54}, Y^{88}, Rh^{102}, W^{181}, W^{185}, and W^{188}-Re^{188}. Radiotungsten contributed the major fraction of the total activity. In suspension studies, chelating agents as compared to water, generally increased the amount of soluble radionuclides, but the effect was small. The effect of water and a chelating agent on the movement of radionuclides in soil columns were also studied.

ACKNOWLEDGEMENT

The authors wish to acknowledge the assistance of Mr. Robert A. Wood in radiochemical analyses.

INTRODUCTION

Among the suggested peaceful uses of nuclear energy is its application to large excavation projects (4). In the event nuclear energy is used for excavation, one must consider the possible hazard of radioactive contamination of the environment. Radionuclides deposited on agriculturally important lands are subject to incorporation into soil and ultimately into the food chain of man via plants and animals. Several laboratories are studying the biological cycling of radioactive fission products dispersed into the biosphere as a result of nuclear weapons testing. Reviews concerning this subject have been presented by Nishita et al. (7), Caldecott (1), and others (2, 8, 9, 11, 12).

This paper is concerned with fallout material from an underground thermonuclear detonation which was designed to study the use of nuclear energy for excavation purposes (Sedan Operation, Nevada Test Site, July, 1962). The objectives of this work were to study the influence of water, chelating agents, and dilute HCl on the release of the radionuclides from the fallout material and the influence of water and a chelating agent on the movement of these nuclides in soil columns.

MATERIALS AND METHODS

The fallout material used in these experiments was collected from an area that was 5400 ft. N 22.5^{o} E of ground-zero. This area corresponded to station number 9 reported by Lane (5).

The release of radionuclides from the fallout material was studied in the following way. One-half gram portions of fallout material were suspended in 25 ml. of distilled H_2O, 0.1 M HCl, 10^{-4}M solutions of sodium-diethylenetriaminepentaacetate (NaDTPA), sodium-cyclohexane-1, 2-diaminetetraacetate (NaCDTA), or sodium-ethylenediamine di

(o-hydroxyphenlacetate) (NaEDDHA) and extracted by centrifuging at approximately 15,000 times gravity at tne end of various time intervals ranging from 5 minutes to 106 days. The supernatant solutions were radioassayed immediately after centrifugation. This procedure was started 33 days after detonation.

To study the movement of soluble radionuclides from the fallout material in soil columns, a 5-g portion of fallout material was mixed with 95 g of either Yolo loam, Hanford sandy loam, or Sorrento loam, and 20 grams of this mixture was placed on top of a column of the same uncontaminated soil used to make the mixture. Some chemical and physical characteristics of these soils and the fallout material are listed in Table 1. The columns (1.5 inches in diameter and containing a total of 300 g of the soil) were leached with a total of 30 inches (870 ml) of distilled water or a solution of CaDTPA (10^{-4}M), which were applied at the rate of 5 inches (145 ml) per irrigation.

At the end of leaching, the soil columns were cut into half-inch increments and radioassayed for total gamma and total beta activity. The solution from each 5-inch leaching was collected and radioassayed. These leaching studies were started 21 days after detonation time.

A 2 x 2 inch thallium-activated sodium iodide crystal-photomultiplier assembly and scaler were used for all the total gamma assays. A mica end-window GM tube (TGC-2, 1.9 mg/cm^2) was used for all beta assays. Radionuclides were identified by gamma ray spectrometry using a 256-channel pulse height analyser with a 3 x 3 inch thallium-activated sodium iodide crystal. Scandium 46, Mn54, Y88, Rh102, Cs137-Ba137, and radiotungsten were confirmed by gamma spectrometry, following radiochemical separations.

RESULTS AND DISCUSSION

Radionuclides Identified

A large number of radionuclides are formed in nuclear detonations. These radionuclides include fission products, neutron-activated nuclides derived from the materials contained in or surrounding the nuclear device (including soil), plus fusion products in the case of thermonuclear detonations. Most of the radionuclides formed are very short lived. Since the present analyses were started 20 days after detonation time, many of the short-lived radionuclides that may have formed were not observed. Table 2 lists the gamma emitting radionuclides positively identified in the fallout material used in the present study. A gamma spectrum of the fallout material 167 days after detonation time is shown in figure 1. The position of the gamma energy peaks of some of the radionuclides that were identified by radiochemical separations or by gamma spectrum analysis are shown. Radiotungsten, presumably formed by neutron-activation, was, by far, the most abundant radionuclide present in the fallout material. The prominence of radiotungsten in the fallout material may be illustrated by referring to figure 1. The contribution of radiotungsten to the activity of the fallout material 167 days after detonation time was estimated to be about 90 per cent of the total gamma activity in the fallout material. This estimate was made by dividing the sum of the radioactivities between the gamma energies of 0.039 and 0.079 Mev., which include the W x-ray peak, by the sum of the radioactivities between 0.039 Mev. and approximately 2.7 Mev. The contributions of backscatter, compton, and bremsstrahlung components to the W x-ray region of the spectrum were not appreciable. The contribution of other radiotungsten isotopes was negligible, that of the next most prominent peak (W181, at 0.152 Mev.)

being less than one per cent. These results support those of Lane (5), who has shown that W187 was one of the more abundant radionuclides 2 days after detonation. Of the tungsten radionuclides contributing to the gamma activity of the samples, W181 apparently was the most abundant. Tungsten 185 was also present in appreciable quantities and was estimated by the absorber technique to be the major beta contributor to the activity of water extracts of the fallout material. Analyses of gamma spectra also indicated the presence of W188-Re188. The term "radiotungsten" as used in this paper includes all of the gamma- or X-ray emitting tungsten isotopes found in the fallout material. The predominance of radiotungsten was also observed in plants that were grown in fallout material.[1]

Soil Column Studies

The gamma activity found at various depths in soil columns that had been leached with distilled water or CaDTPA solution is shown in figure 2. The gamma activity at various depths is expressed as a fraction of the total activity applied to the soil column. In all cases the fraction of beta activity, which is not shown, corresponded very closely with the fraction of gamma activity. More radioactivity was found in the CaDTPA leachate than in the water leachate, which indicated that the CaDTPA solution was the more effective leaching agent. Gamma ray spectra of the leachate solutions from Yolo soil (fig. 3) indicated that the H_2O leachate contained predominantly I131 and some Ru103 and K40. In addition to these radionuclides, the CaDTPA leachates contained Ce141 and La140. Lanthanum 140 was present but the parent Ba140 was not observed. Thus, La140 apparently was complexed and leached from the soil by DTPA, but the parent Ba140 was retained in the soil. The gamma spectra of leachates from Sorrento soil were similar to those of Yolo except that the amount of La140 was much lower. By comparing the 0.364 Mev. gamma

[1] E. M. Romney, private communication

peak of I131 for both H_2O and CaDTPA leachates it was conjectured that I131 movement was not affected by the chelate treatment. To confirm this, a separate experiment was conducted. In this experiment, columns of Yolo and Sorrento soil contaminated with carrier-free I131 were leached with distilled H_2O or CaDTPA solution. Although the two soils differed in the retention of I131, no chelate effect on the movement of I131 was observed.

Under the conditions of the experiments, the effect of CaDTPA in altering the gamma activity of the leachate solutions was only slightly greater than that of H_2O. Of greater significance was the effect of soil type on the movement of radionuclides from the initial zone of contamination (Table 3). The greatest amount of radionuclide movement occurred in Sorrento and the least in Hanford. Table 4 lists the amounts of radiotungsten found at various depths, represented as fraction of total gamma activity applied to the soil column. The total gamma activity applied can be obtained from the decay curve shown in figure 4. By comparing the data of column A, Table 4, with those of figure 2, which shows the total gamma activities, it can be seen that the radioactivity found to a depth of 6.0, 4.5, and 2.3 inches in Yolo, Sorrento and Hanford soils, respectively, was due predominantly to radiotungsten. This is also shown in column B of Table 4. More radiotungsten moved in Yolo (pH 8) and Sorrento (pH 7.8) soils than in Hanford (pH 6.6). This difference among the soils indicated that the radiotungsten movement may depend somewhat on soil pH. These results agree with the chemical properties of tungsten, which reacts as an anion and is more soluble in aqueous solutions of higher pH containing alkali metal ions than in acidic solutions (6, 10). In a suspension experiment involving the use of clay systems, W185 sorption was found to

depend on clay mineral type[1]. Kaolinite and illite sorbed large amounts of W185, whereas bentonite sorbed relatively small amounts. Since Hanford soil contains predominantly illite, Sorrento contains kaolinite with some montmorillonite, and Yolo contains montmorillonite with some kaolinite, the degree of radiotungsten movement in these soils may be directly related to the sorption characteristics of the predominant clay mineral.

The amount of radioactivity found in leachate solutions in relation to the number of leachings is shown in figure 5. Except for the CaDTPA treatment of Hanford, an apparent steady-state removal of radionuclides appeared after the third leaching both with water and CaDTPA. This was attributed to the limited water solubility of the throwout material.

Suspension Studies

The degree of dissolution of the fallout material in H_2O, HCl, DTPA, CDTA, and EDDHA solutions was investigated by the suspension method. Fallout material was suspended in these compounds for periods of time ranging from 5 minutes to 106 days. Figure 6 shows the amount of radioactivity found in the supernatant liquid at various time intervals. All treatments decreased the amount of radioactivity in the supernatant liquid between 5 minutes and 30 minutes (between 5 minutes and 2 hours for H_2O). The reduction of this initially high radioactivity may have occurred by increased sorption of the soluble radionuclides on soil colloids and by decay of short lived radionuclides. With all treatments, except HCl, an increase of radioactivity in the supernatant liquid was observed between 30 minutes (2 hours for H_2O) and 7 days. Continued dissolution of the fallout material and accumulation of soluble radiotungsten in the supernatant liquid, which had a pH of 10, accounts for the increase in radioactivity with time. After 7 days the amount of radioactivity in the

supernatant liquid decreased. This observed decrease may be explained by the following considerations. The rate of dissolution of the fallout material may have decreased as suspension time progressed. The gross rate of radionuclide decay may then have exceeded the rate at which radionuclides were being dissolved from the fallout material.

Hydrochloric acid initially dissolved more radionuclide than did H_2O, and gave a similar decrease in radioactivity recovered. However, the amount of radioactivity recovered continued to decrease throughout the entire experiment. This continued decrease was attributed to sorption on soil colloids and decay of soluble radionuclides and to the continued removal of a large portion of radiotungsten from the HCl supernatant liquid.

Chelating agents increased the total gamma activity of the supernatant liquids over that of H_2O; however, the increase was not large. The order of effectiveness of chelating agents in increasing gamma activity between 30 minutes and 7 days was: EDDHA>DTPA>CDTA>H_2O, while after 65 days the order of effect was: CDTA>EDDHA>DTPA>H_2O.

Portions of gamma ray spectra of the 106 day supernatant liquid are presented in figure 7. Except for increased Ru103-106 and the appearance of Mn54, chelating agents caused the dissolution of nearly the same amounts of radionuclides as did H_2O. The most abundant radionuclide was radiotungsten for H_2O, DTPA, CDTA, and EDDHA supernatant liquids; however, besides radiotungsten, considerable amounts of Ce141-144, and Ru103-106 were also found in the HCl supernatant liquids. Hydrochloric acid drastically reduced the amount of radiotungsten but increased the amount of Ce141-144 and Ru103-106. Manganese 54 was also

present in the HCl supernatant liquids.

SUMMARY

Fallout material from an underground thermonuclear detonation was analyzed to determine the presence of several longer lived radionuclides. Fission products that were identified were: Zr95-Nb95, Ru103, Ru106-Rh106, I131, Cs137-Ba137, Ba140-La140, Ce141, and Ce144-Pr144. Other identified radionuclides, which probably were produced by neutron activation, were: Sc46, Mn54, Y88, Rh102, W181, W185, and W188-Re188. Radiotungsten was estimated to contribute about 90 per cent of the total gamma activity present in the fallout material 167 days after detonation time. The release of the radionuclides from the fallout material was studied in H_2O, chelating agents, and HCl. In H_2O suspensions of fallout material the amount of soluble radionuclides increased up to 7 days whereas in HCl, they decreased. This effect was attributed to the reduced solubility of radiotungsten in acid solutions. Chelating agents as compared to water, generally increased the amount of soluble radionuclides, but the effect was small.

In soil column studies, distilled water or a solution of CaDTPA was used as a leaching agent. Ruthenium 103 and I131 were found in the leachates from the water leached soils. In the leachates from the CaDTPA leached soils, La140 and Ce141 were found in additon to Ru103 and I131. Apparently the complexing action of DTPA increased the solubility of La140 and Ce141. The movement of radiotungsten in soil columns depended on the soil type. Its movement was greater in Yolo (pH 8.0) and Sorrento (pH 7.8) containing montmorillonite than in illitic Hanford soil (pH 6.6).

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Table 1. Some physical and chemical properties of soils and fallout material

	Sorrento	Yolo	Hanford	Fallout Material
pH (1:1 suspension)	7.8	7.9	6.6	9.8
Organic matter (%)	1.85	1.73	1.24	0.16
Cation exchange capacity (me/100g.)	15.78	18.18	4.95	24.26
Extractable cations* (me/100g.)				
Ca	26.9	11.2	4.47	67.6
Mg	1.70	7.40	0.78	11.9
Na	0.045	0.222	0.15	3.10
K	0.122	0.569	0.39	4.90
Carbonate (me/100g.)	18.2	1.61	----	226
Particle size (%)				
Sand	52.8	53.0	67.8	37.8
Silt	35.2	31.6	24.4	51.8
Clay	12.1	17.0	7.85	10.3
Predominant clay mineral**	K + M	M + K	I	M

* Extractable cations with neutral N ammonium acetate

** M + K = Montmorillonite plus some Kaolinite
K + M = Kaolinite plus some Montmorillonite
I = Illite

Table 2. Radionuclides identified in the fallout material.

Nuclide	Half-life		Probable Mode of Formation	Gamma Transitions	Reference
Fission Products				Mev.	
40Zr95	65	d*	F**	0.722, 0.754	13
↓ 41Nb95	35	d		0.765	
44Ru103	39.8	d	F	0.498, 0.610	13
↓ 45Rh103^{m}	57	m			
44Ru106	365	d	F	no γ	13
↓ 45Rh106	30	s		0.513, 0.624	
53I131	8.08	d	F	0.163, 0.284, 0.364 0.673, 0.722	13
55Cs137	33	y	F		
↓ 56Ba137^{m}	2.60	m		0.662	13
56Ba140	12.8	d	F	0.132, 0.162, 0.304 0.436, 0.537	13
↓ 57La140	40.2	h		0.329, 0.431, 0.486, 0.752, 0.816, 0.926, 1.60, 1.90, 2.55	
58Ce141	33.1	d	F	0.142	13
58Ce144	285	d	F	0.134	13
↓ 59Pr144	17.3	m		0.696, 1.48, 2.18	
Neutron-induced					
21Sc46	83.9	d	(n, γ)Sc45	0.885, 1.12	13
25Mn54	291	d	(n, 2n)Mn55	0.840	13
39Y88	104	d	(n, 2n)Y89	0.908, 1.85, 2.76	13
45Rh102	210	d	(n, 2n)Rh103	0.125, 0.200, 0.475 0.635, 0.72, 0.79, 1.08	13
74W181	145	d	(n, γ)W180	0.136, 0.152, EC, Ta-K_{α} x-ray 0.058	3, 13

Table 2. (Cont.)

Nuclide	Half-life	Probable Mode of Formation	Gamma Transitions	Reference
Neutron-induced (cont'd)			Mev.	
74W185	75.8 d	(n, γ)W184 (n, 2n)W186	Re-K_{α} x-ray 0.060 ($\beta^-_{E\ max}$ = 0.43 Mev.	3, 13 13
74W188 ↘ 75Re188	69.5 d 16.7 h	(n, γ)W187+	? 0.155	
Naturally radioactive				
19K40	1.25 x 10^9 y		1.46	13

* s = second; m = minute; h = hour; d = day; y = year

**Direct or decendant of fission product.

+ Second order activation (W187 first produced by neutron activation).

Table 3. Fraction of total gamma activities found in the leachate and in the soil columns below the initial contamination zone.

	H_2O*			CaDTPA*		
Soil	Fraction in Soil Column	Fraction in Leachate	Sum	Fraction in Soil Column	Fraction in Leachate	Sum
Sorrento	0.184	0.0134	0.197	0.212	0.0231	0.235
Yolo	0.108	0.0128	0.121	0.181	0.0250	0.206
Hanford	0.115	0.0114	0.126	0.088	0.0135	0.102

*Fraction of total gamma activity applied in the form of fallout material.

Table 4. Radiotungsten activity found at various depths in soil columns leached with distilled water and CaDTPA.

Sorrento			Yolo			Hanford		
Depth inches	A	B	Depth inches	A	B	Depth inches	A	B
			H_2O Leaching					
0.47	0.36	0.47	0.16	0.24	0.59	0.29	0.42	0.54
1.20	0.039	0.79	0.61	0.25	0.63	0.84	0.064	0.86
1.71	0.033	0.79	1.15	0.038	1.00	1.35	0.018	0.84
2.20	0.030	0.76	1.69	0.017	0.99	1.84	0.0021	0.83
2.74	0.021	0.84	2.23	0.013	0.97	2.32	trace	----
3.24	0.010	0.77	2.75	0.011	1.05			
3.74	0.0048	0.77	3.28	0.0075	0.92			
4.23	0.0015	0.75	3.80	0.0045	0.98			
4.70	trace	----	4.31	0.0029	1.04			
			4.84	0.0014	0.86			
			5.36	0.0006	0.76			
			5.89	trace				
			CaDTPA Leaching					
0.38	0.36	0.45	0.36	0.40	0.54	0.30	0.47	0.52
1.02	0.074	0.90	0.98	0.085	0.87	0.84	0.072	0.95
1.53	0.042	0.97	1.49	0.023	0.88	1.33	0.010	0.96
2.03	0.035	0.93	2.00	0.015	0.96	1.84	0.0025	0.94
2.54	0.023	0.86	2.49	0.0088	0.82	2.35	trace	----
3.04	0.014	0.97	2.98	0.0061	0.86			
3.54	0.0061	0.90	3.47	0.0049	1.04			
4.04	0.0023	0.96	3.98	0.0025	0.88			
4.56	trace	----	4.48	0.0017	1.05			
			4.98	0.0008	0.95			
			5.48	0.0005	0.90			
			5.98	trace	----			

A. Radiotungsten activity expressed as the fraction of total gamma activity applied to the soil column.

B. Radiotungsten activity expressed as the fraction of total gamma activity at each depth.

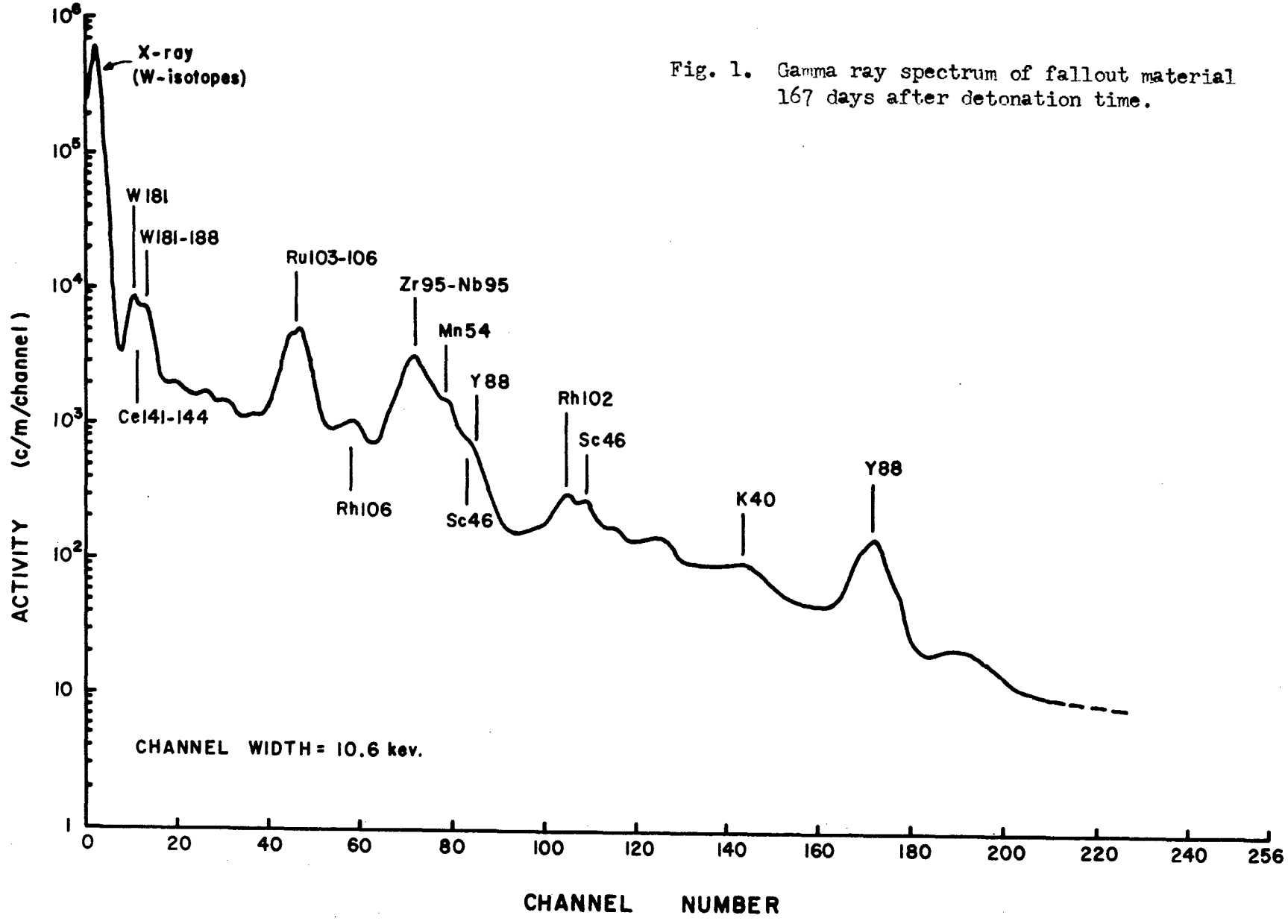

Fig. 1. Gamma ray spectrum of fallout material 167 days after detonation time.

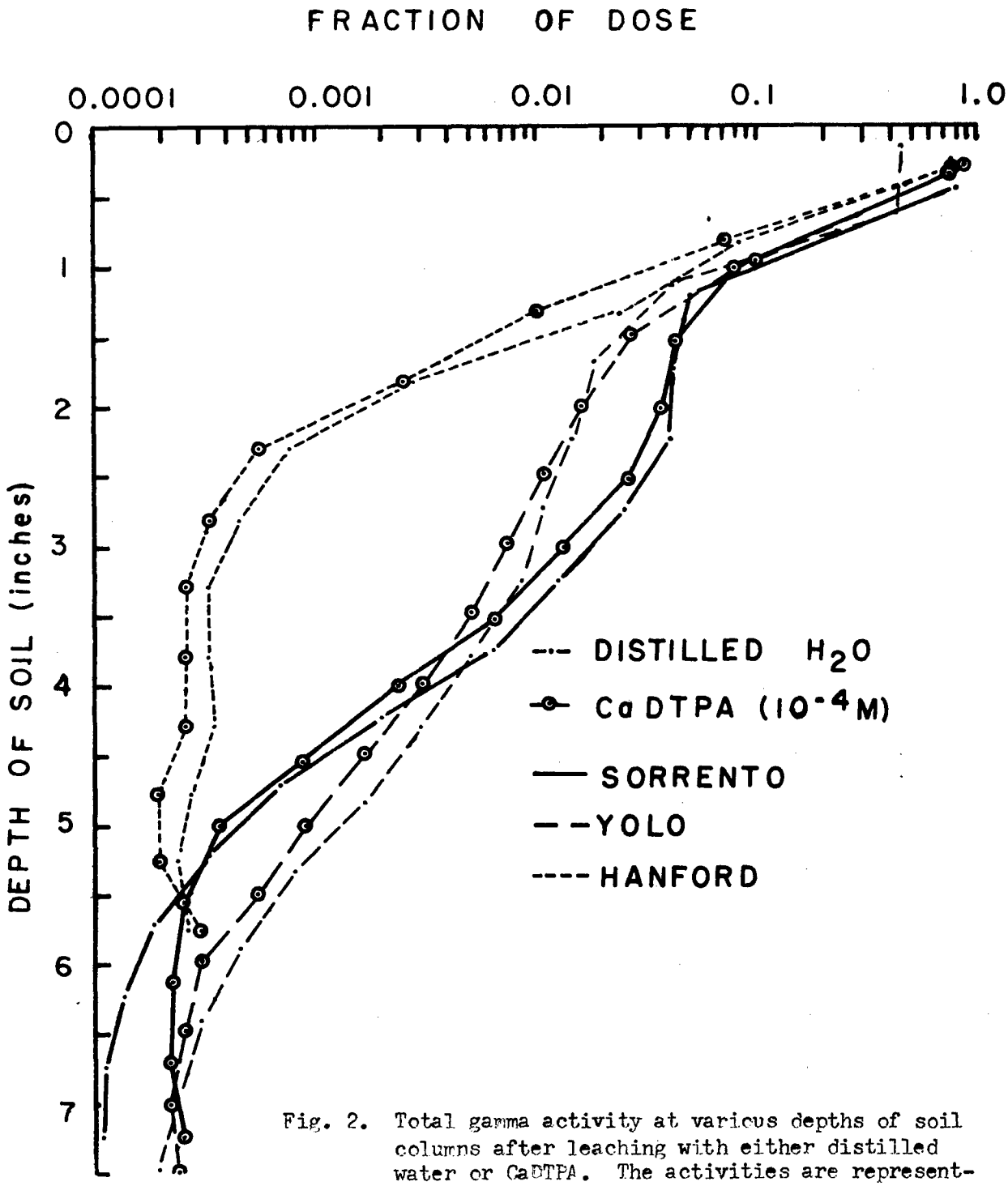

Fig. 2. Total gamma activity at various depths of soil columns after leaching with either distilled water or CaDTPA. The activities are represented as fraction of total activity applied to the soil column in the form of fallout material.

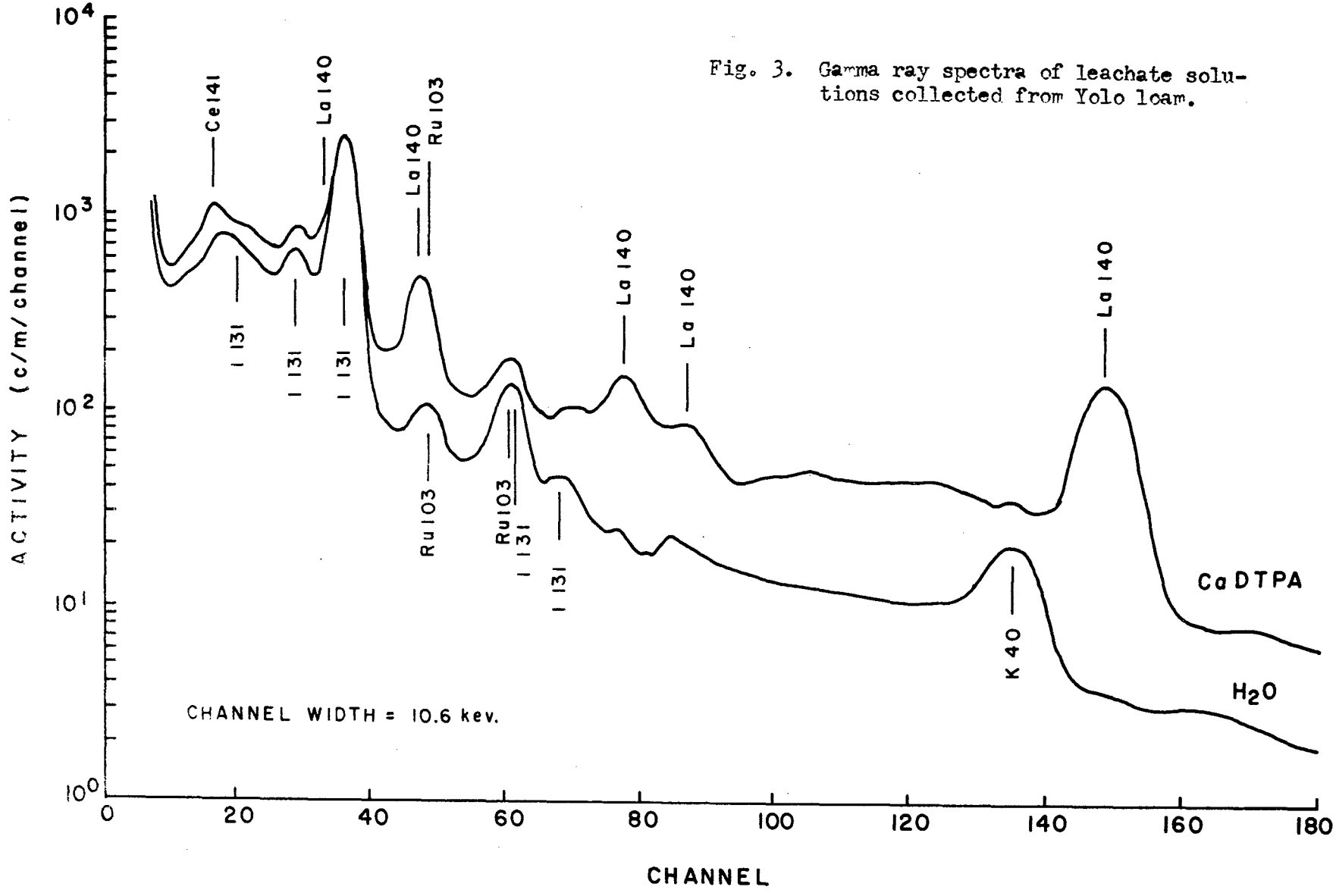

Fig. 3. Gamma ray spectra of leachate solutions collected from Yolo loam.

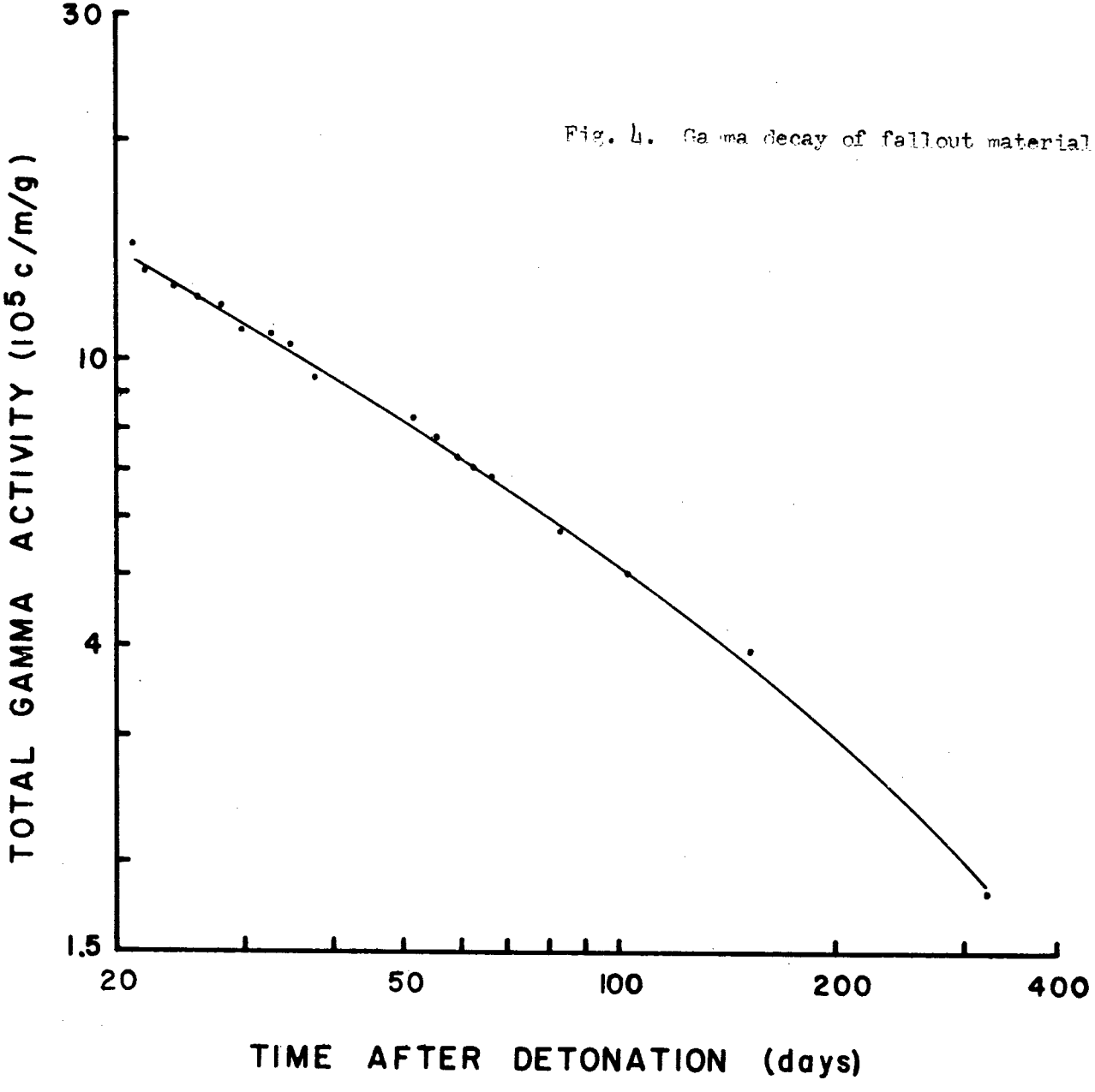

Fig. 4. Gamma decay of fallout material

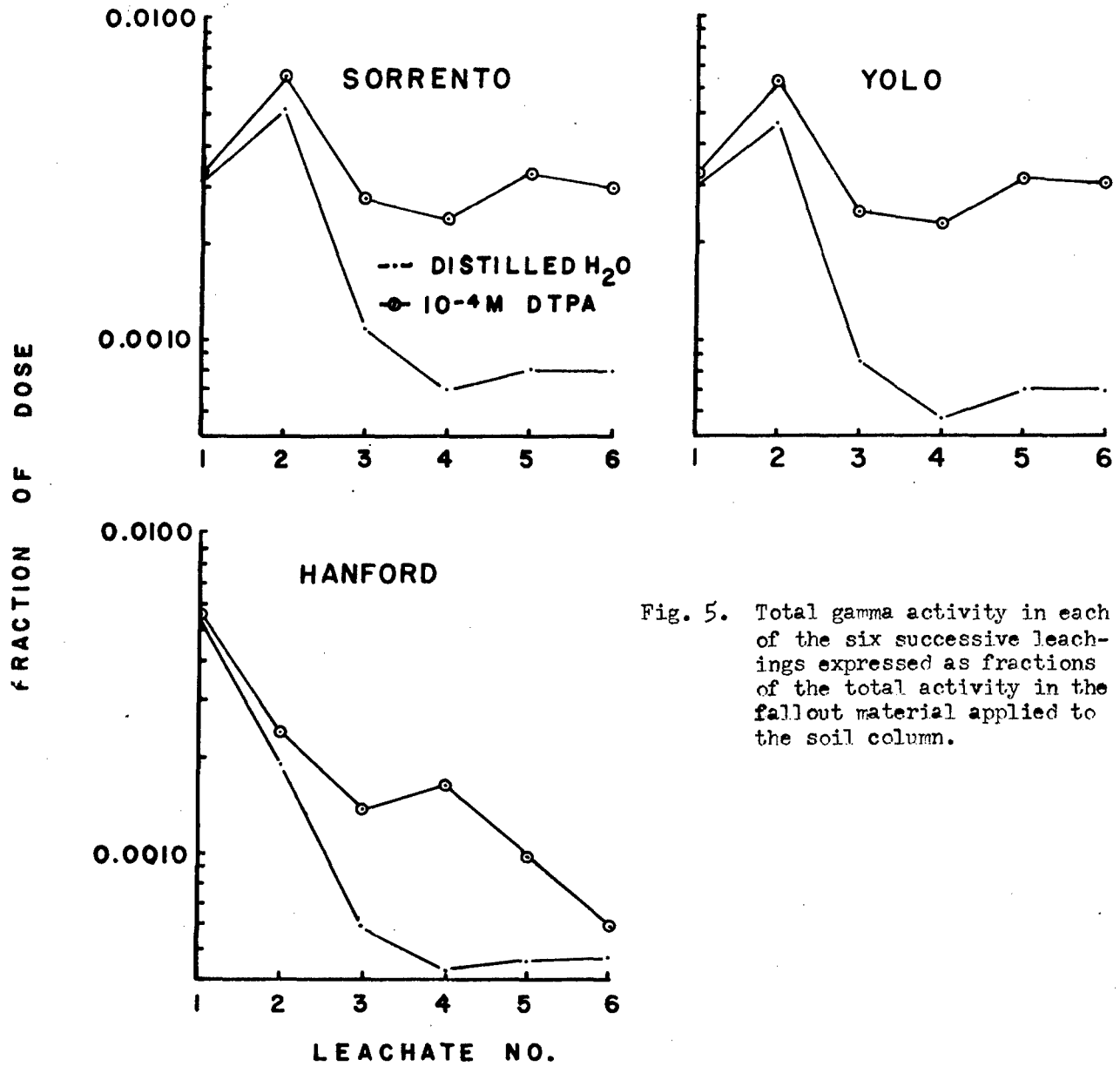

Fig. 5. Total gamma activity in each of the six successive leachings expressed as fractions of the total activity in the fallout material applied to the soil column.

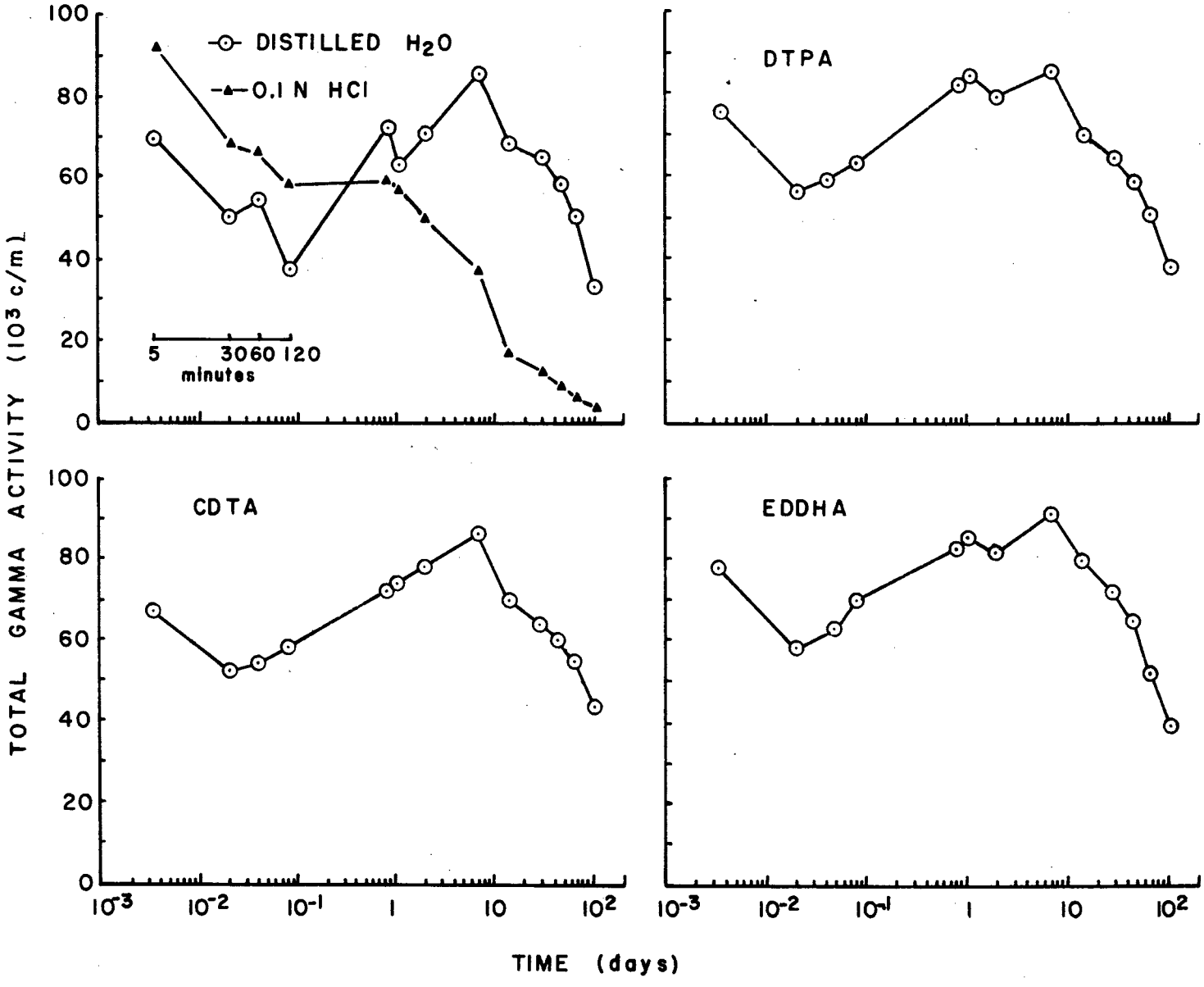

Fig. 6. Total gamma activity of supernatant liquid from suspensions of fallout material and H_2O, HCl, DTPA, CDTA, or EDDHA.

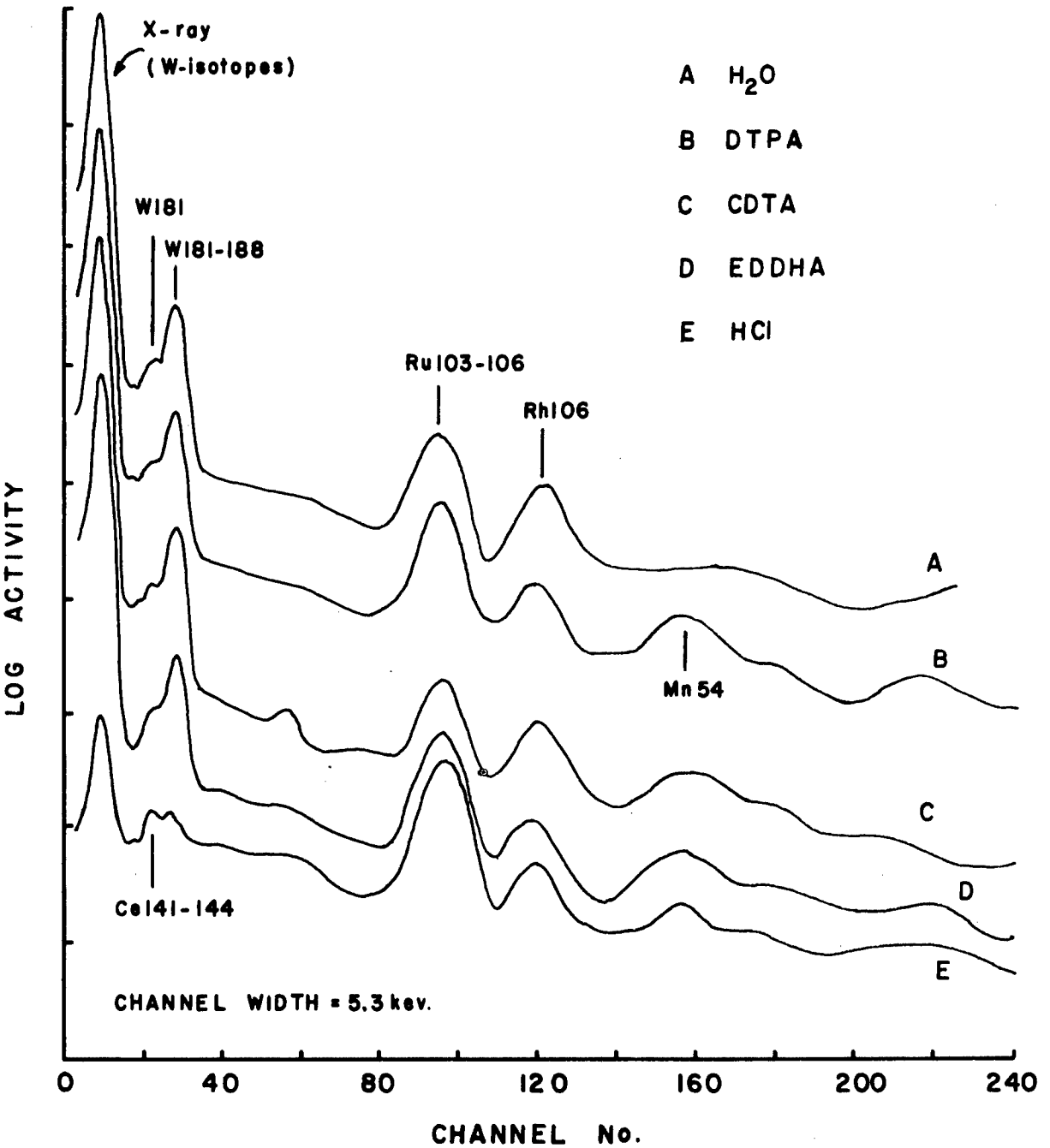

Figure 7.

Portions of the Gamma Ray Activity in 106 Day Supernatant Liquid.

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LRL	231F	Rad-Chem Analysis
LRL	232P	Yield Measurements
EGG	233P	Timing and Firing
WES	234P	Stability of Cratered Slopes
LRL	235F	Seismic Velocity Studies

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USC-GS	213P	"Seismic Effects From a High Yield Nuclear Cratering Experiment in Desert Alluvium"
NRDL	229P	"Some Radiochemical and Physical Measurements of Debris from an Underground Nuclear Explosion"
NRDL	230P	Naval Aerial Photographic Analysis

ABBREVIATIONS FOR TECHNICAL AGENCIES

STL	Space Technology Laboratories, Inc., Redondo Beach, Calif.
SC	Sandia Corporation, Sandia Base, Albuquerque, New Mexico
USC&GS	U. S. Coast and Geodetic Survey, San Francisco, California
LRL	Lawrence Radiation Laboratory, Livermore, California
LRL-N	Lawrence Radiation Laboratory, Mercury, Nevada
Boeing	The Boeing Company, Aero-Space Division, Seattle 24, Washington
USGS	Geological Survey, Denver, Colorado, Menlo Park, Calif., and Vicksburg, Mississippi
WES	USA Corps of Engineers, Waterways Experiment Station, Jackson, Mississippi
EGG	Edgerton, Germeshausen, and Grier, Inc., Las Vegas, Nevada, Santa Barbara, Calif., and Boston, Massachusetts
BYU	Brigham Young University, Provo, Utah
UCLA	UCLA School of Medicine, Dept. of Biophysics and Nuclear Medicine, Los Angeles, Calif.
NRDL	Naval Radiological Defense Laboratory, Hunters Point, Calif.
USPHS	U. S. Public Health Service, Las Vegas, Nevada
USWB	U. S. Weather Bureau, Las Vegas, Nevada
USBM	U. S. Bureau of Mines, Washington, D. C.
FAA	Federal Aviation Agency, Salt Lake City, Utah
REECO	Reynolds Electrical and Engineering Co., Las Vegas, Nevada

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